Determination of Actinide and Fission Product Isotopes in High-Burnup Spent Nuclear Fuel

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Introduction

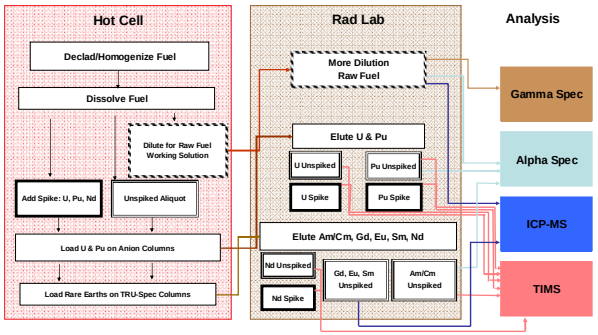
- Interest among nuclear power producers has grown over the past few decades in higher utilization of nuclear fuel, which translates to achieving higher burnup.
- Argonne's Energy Technology Division (ET) has worked with the Nuclear Regulatory Commission (NRC) for several years to investigate materials issues related to storage of high-burnup fuel, including studies of hydrogen migration in cladding, thermal creep, and mechanical performance.
- The Chemical Engineering Division (CMT) has undertaken the task of analyzing specimens of fuel from rods used in the ET studies to determine actinide and fission product isotopes.
  - An analysis scheme that can produce data for about 75 isotopes from a small number of operations has been applied to two high-burnup specimens (>70 GWd/MTU burnup).
  - The primary interest of NRC in these analyses was for isotopes needed to assign an experimental burnup value to the fuels, and for isotopes related to burnup credit considerations. The analysis goals were extended to include isotopes that affect heat-load, shielding, and criticality considerations.

High-Burnup Fuels

- Burnup is a measure of the number of fuel atoms that underwent fission.
- Of interest to the NRC is determining the reliability of existing computational codes for predicting nuclide inventories at burnup levels beyond those that are already validated (to about 40 GWd/MtU).
- Burnup credit is an allowance in safety analysis calculations for decreased reactivity in the fuel as a result of actinide depletion and a presence of neutron-absorbing irradiation products (poisons) that reduce the fuel's ability to achieve criticality.
- At high burnup, the decrease in reactivity can be substantial and might allow significant increases in the quantity of spent fuel that can be safely stored or transported in a single container, which could reduce storage and transportation costs.

Experimental data for isotopes in fuels with burnup higher than 40 GWd/MtU are limited. Data for some isotopes that are important to burnup-credit calculations are very rare.

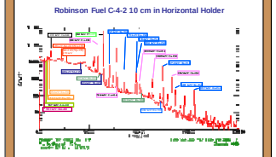
Operations Flowsheet



Gamma Spectrometry

- Semiconductor detector and multi-channel analyzer
- Records gamma counts vs. energy; precision depends on count rate

^{134}Cs, ^{137}Cs, ^{106}Ru/Rh, $^{154/155}$Eu, ^{241}Am, ^{144}Ce, ^{125}Sb



Thermal Ionization Mass Spectrometry (TIMS)

- Conventional high-precision solid-source, magnetic-sector
- Isotope ratios with < 0.1% RSD for major isotopes
- Assay data using isotope dilution

U, Pu, Nd isotopes for Burnup

^{243}Am, ^{245}Cm, ^{246}Cm, ^{247}Cm

Mass Fraction in Fuel, g/g		
	U	Pu
C-4 #1	0.7615	0.010045
C-4 #2	0.7604	0.010095
% RSD	**0.10**	**0.35**
C-22 #1	0.7738	0.010492
C-22 #2	0.7752	0.010502
% RSD	**0.13**	**0.07**

Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

- RF-excited argon plasma source with quadrupole mass analyzer
- Records ions vs. mass; suffers from "isobaric interferences" (same mass, different elements)

Multi-element quantitation (5% RSD):

Most Burnup-Credit Fission Products

^{88}Sr, ^{89}Y, ^{91}Zr, ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{109}Ag, ^{111}Cd, $^{133/135}$Cs, ^{138}Ba, ^{139}La, ^{140}Ce, ^{141}Pr, $^{143/145}$Nd, $^{149/152}$Sm, ^{153}Eu, $^{156/157/158/160}$Gd, ^{237}Np

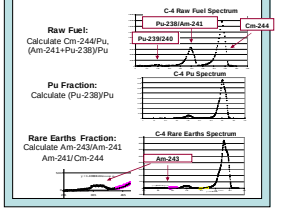
Isotope Ratio after separations (1% RSD for majors):

$^{144\text{-}152/154}$Sm, $^{150\text{-}155}$Eu, $^{150,152\text{-}158,160}$Gd

Alpha Pulse Height Analysis (Alpha Spectrometry)

- Semiconductor detector and multi-channel analyzer
- Records alpha counts vs. energy; precision depends on count rate

^{238}Pu, ^{241}Am, ^{244}Cm, (^{243}Am)



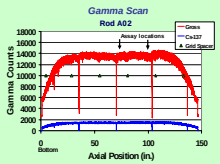
Axial Variations in Parent Fuel Rod

Very little difference was found between the isotope concentrations in the two fuel specimens (C4 and C22).

This was expected based on the gamma scan of the fuel rod before sampling.

Gamma Analysis – µCi/g of Fuel

	C4	C22
^{137}Cs	$1.31x10^{5}$	$1.38x10^{5}$
^{134}Cs	$1.57x10^{4}$	$1.69x10^{4}$
^{106}Ru	$1.34x10^{3}$	$1.32x10^{3}$
^{125}Sb	$5.57x10^{2}$	$6.07x10^{2}$
^{144}Ce	$2.53x10^{2}$	$2.86x10^{2}$
^{154}Eu	$5.24x10^{3}$	$5.45x10^{3}$
^{155}Eu	$2.18x10^{3}$	$2.41x10^{3}$
^{241}Am	$1.99x10^{3}$	$2.07x10^{3}$



C4 ~15 mm below fuel midplane; in grid span #3
C22 ~750 mm above fuel midplane; in grid span #4

Conclusions

Many data were collected in relatively few measurements

- 75 isotopes were measured
- ICP-MS is especially productive in spent fuel analysis when moderate accuracy is tolerable and isobaric interferences are dealt with

Noteworthy data were collected for burnup credit isotopes

- 19 of 22 ranked fission product isotopes reported
- Measurements were made of Rh-103, Mo-95, and Ru-101, isotopes for which measured data are very rare.

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